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ROLE OF NNN INTERLAYER COUPLING IN BILAYER GRAPHENE

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Abstract: The present work deals with the role of next-nearest-neighbour (NNN) interlayer coupling parameter (tp') on the electronic structure of AA-stacked bilayer graphene (AA-BLG). Hereby, considering the tight-binding Hamiltonian with NNN interlayer coupling and employing the green function method to determine the parameters such as quasi-particle energy and density of states for AA-BLG. It will also assess how these parameters affect their electronic properties by analyzing variations in band structure, density of states and conductivity. Results show that on increasing the value of tp', the gap between the valence and the conduction band decreases. The density of state (DOS) graph points toward the splitting as well as broadening of quasi-particle peaks with increasing values tp', the quasi-particle peak of the conduction band increases and that of the valence band decreases, signifying transfer of quasi-particles from valence to the conduction band.

Keywords: AA-stacked bilayer graphene, Tight-Binding Hamiltonian, Band structure, Tunable bandgap.

1. INTRODUCTION

Graphene's isolation by K. S. Novoselov and A. K. Geim [1] in 2004–2005 has revolutionized scientist's understanding of two-dimensional materials, revealing extraordinary electronic properties characterized by massless Dirac fermions [2]. Single-layer graphene (SLG) exhibits a linear energy dispersion near the Dirac points [3,5-7]. However, when layers are stacked, interlayer interactions can drastically modify electronic behaviour. Among the different stacking arrangements, AA-stacked bilayer graphene (AA-BLG) is particularly intriguing due to its perfect atomic alignment, which preserves much of the intrinsic linear dispersion observed in monolayer graphene, yet introduces unique features in its quasi-particle spectrum and density of states (DOS) [4,7]. Early theoretical work laid the foundation for understanding these interlayer interactions. For instance, tight-binding models developed by Charlier et al. [8, 9] provided critical insights into the electronic structure of graphite and graphene. Subsequent studies, such as those by McCann and Koshino [4], have further elucidated the role of nearest-neighbour (NN) and next-nearest-neighbour (NNN) interlayer couplings, showing that NN coupling in AB-stacked graphene leads to significant band splitting and tunable band gaps.

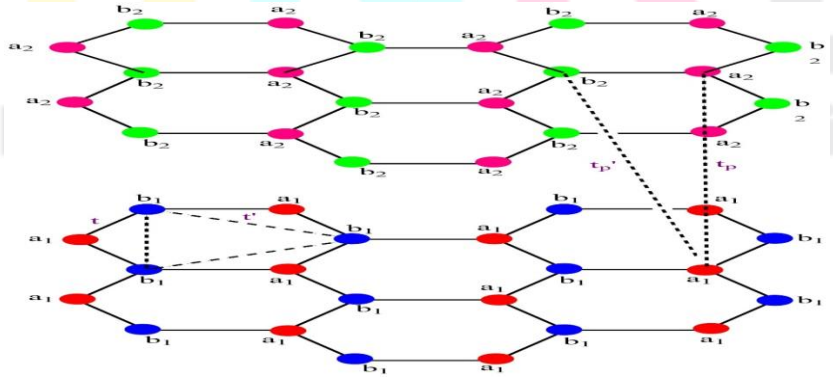

Fig.1 Atomic structure of AA-stacked bilayer graphene.

In contrast, AA stacking maintains more symmetric electronic features. Additionally, investigations by Zhang et al. [10] and Ohta et al. [11, 12] have highlighted how subtle variations in interlayer hopping parameters can influence optical and transport properties. These collective studies underscore the importance of a detailed understanding of interlayer coupling parameters for tailoring the properties of bilayer graphene for nanoelectronics' applications. In this work, we focus on AA-stacked bilayer graphene to explore how its unique stacking order, governed by NN and NNN interlayer interactions, affects its quasi-particle

energy spectrum and DOS. By systematically analysing these effects, our study aims to advance the fundamental understanding of interlayer coupling in graphene and guide the optimization of its electronic properties for practical device applications.

2. THEORETICAL METHOD

Tight-Binding Hamiltonian for AA-stacked bilayer graphene within two triangular sub-lattice approach in momentum space is divided into two parts, i.e. Hamiltonian for intralayer and interlayer. Considering t as nearest-neighbour hopping parameter and t' as next-nearest-neighbour hopping parameter on triangular sub-lattices [2,3,4]. Also, t_p is the nearest-neighbour [16] and t'_p is the interlayer coupling parameter for next-nearest-neighbour. Now the Hamiltonian for AA-BLG is written below;

$$H_{AA-Bilayer}=H_{Intralayer}+H_{Interlayer} \quad (1)$$

$$H_{Intralayer} = -\sum_{k,\sigma,\alpha=1,2} t_k\{\, a^{+}_{\alpha k\sigma}b_{\alpha k\sigma} + h.c.\}-\sum_{k,\sigma,\alpha=1,2} t'_k\{a^{+}_{\alpha k\sigma}a_{\alpha k\sigma} + b^{+}_{\alpha k\sigma}b_{\alpha k\sigma} + h.c.\} \quad (1a)$$

$$H_{Interlayer}= -\sum_{\substack{k,\sigma,\alpha,\beta \\ \alpha\neq\beta}} t_p\{\, a^{+}_{\alpha k\sigma}a_{\beta k\sigma} + b^{+}_{\alpha k\sigma}b_{\beta k\sigma} + h.c.\} -\sum_{\substack{k,\sigma,\alpha,\beta \\ \alpha\neq\beta}} t'_p\{a^{+}_{\alpha k\sigma}b_{\beta k\sigma} + b^{+}_{\alpha k\sigma}a_{\beta k\sigma} + h.c.\} \quad (1b)$$

where, α and β are layer index. The $a^{+}_{\alpha k\sigma}(b_{\alpha k\sigma})$ creates (annihilates) an electron on sub-lattice A(B) with momentum (k) and spin (σ). Also, $t_k = t\{3+f(k)\}^{\frac{1}{2}}$; $t'_k = -t'\{3+f(k)\}^{\frac{1}{2}}$ and $f(k) = 2\cos(\sqrt{3}k_y) + 4\cos\left(\frac{\sqrt{3}}{2}k_y\right)\cos\left(\frac{3}{2}k_x\right)$ (2)

And we have assumed Fermi energy (E_F) at 0eV. We assumed a green function, to study the nature of quasi-particles and electronic states in AA-BLG i.e. given by; $G^{11}_{AA}(k,\omega) = \ll a_{1k\sigma}/a^{+}_{1k\sigma} \gg$. With the help of above tight binding Hamiltonian and green function we get the four equations of motion for AA-BLG:

$$(\omega+2t'_k) \ll a_{1k\sigma}, a^{+}_{1k\sigma} \gg = \frac{1}{2\pi} - t_k \ll b_{1k\sigma}, a^{+}_{1k\sigma} \gg - t_p \ll a_{2k\sigma}, a^{+}_{1k\sigma} \gg - t'_p \ll b_{2k\sigma}, a^{+}_{1k\sigma} \gg \quad (3)$$

$$(\omega+2t'_k) \ll b_{1k\sigma}, a^{+}_{1k\sigma} \gg = - t_k \ll a_{1k\sigma}, a^{+}_{1k\sigma} \gg - t_p \ll b_{2k\sigma}, a^{+}_{1k\sigma} \gg - t'_p \ll a_{2k\sigma}, a^{+}_{1k\sigma} \gg \quad (4)$$

$$(\omega+2t'_k) \ll a_{2k\sigma}, a^{+}_{1k\sigma} \gg = -t_k \ll b_{2k\sigma}, a^{+}_{1k\sigma} \gg - t_p \ll a_{1k\sigma}, a^{+}_{1k\sigma} \gg - t'_p \ll b_{1k\sigma}, a^{+}_{1k\sigma} \gg \quad (5)$$

$$(\omega+2t'_k) \ll b_{2k\sigma}, a^{+}_{1k\sigma} \gg = -t_k \ll a_{2k\sigma}, a^{+}_{1k\sigma} \gg - t_p \ll b_{1k\sigma}, a^{+}_{1k\sigma} \gg - t'_p \ll a_{1k\sigma}, a^{+}_{1k\sigma} \gg \quad (6)$$

After solving the above equations (3)-(6), we obtained the value of the desirable green function i.e.

$$G^{11}_{AA}(k,\omega) = \ll a_{1k\sigma}/a^{+}_{1k\sigma} \gg \text{ as; } G^{11}_{AA}(k,\omega) = \frac{1}{2\pi}\left(\frac{A_{ik}}{(\omega-\mu_{1k})(\omega-\mu_{2k})(\omega-\mu_{3k})(\omega-\mu_{4k})}\right) \quad (7)$$

We can rewrite the above green's function, using simple algebra, in the following standard form;

$$G^{11}_{AA}(k,\omega) = \frac{1}{2\pi}\sum_{i=1}^{4}\frac{A_{ik}}{(\omega-\mu_{ik})} \quad (8)$$

where, $A_{ik} = \omega^3 + 6\omega^2 t'_k + \omega\left(-t_k^2 + 12t_k'^2 - t_p'^2 - t_p^2\right) + 2t_p t_k t'_p - 2t'_k t_p^2 - 2t'_k t_k^2 + 8t_k'^3 - 2t'_k t_p'^2$ Where $\omega_{1k}, \omega_{2k}, \omega_{3k}$ and ω_{4k} are the four quasi particle energy branches corresponding to four poles of the above Green's function equation (8) for AA-BLG and given as follows:

$$\mu_{1k} = -2t'_k + t_k + t'_p - t_p;\ \mu_{2k} = -2t'_k - t_k + t'_p + t_p;\ \mu_{3k} = -2t'_k - t_k - t'_p - t_p;\ \mu_{4k} = -2t'_k + t_k - t'_p + t_p \quad (9)$$

above four equations give the dispersion relation, which shows the quasi-particles energy dependence on the NN and NNN interlayer coupling parameter i.e. t_p and t'_p respectively. Equation (9) is important for to analyze the nature of quasi particle energies in AA-BLG. Detailed numerical analysis of the quasi-particle dispersion is given in results and discussion section. Spectral function of $G^{11}_{AA}(k,w)$ can be calculated using eq. (8) by considering the imaginary part of the green function and hence the electronic density of states in the AA-BLG structure. The spectral function for AA-BLG can be written as-

$$A^{11}_{AA}(k,\omega) = -\frac{1}{\pi}\mathrm{Im}G^{11}_{AA}(k,\omega) \quad (10)$$

where, Im stands for imaginary part of Green's function and the final expression for DOS for AA-BLG in terms of spectral function is given as:

$$N_{AA}(\omega) = \iint_{-\pi}^{\pi} A^{11}_{AA}(k,\omega)\, dk_x dk_y \quad (11)$$

where, $A^{11}_{AA}(k,\omega) = \frac{1}{\pi}\sum_{i=1}^{4} A_{ik}\delta(\omega-\mu_{ik})$,

δ-function which is equivalent to broadening the quasi-particle peak and one need to solve δ-function. For this purpose, we considered Lorentzian type of broadening by using the relationship:

$$\delta(\omega-\mu_{ik}) \approx \frac{1}{\pi}\lim_{\Gamma\to 0}\left[\frac{\Gamma}{\Gamma^2+(\omega-\mu_{ik})^2}\right]$$

where Γ is broadening factor independent of k and ω for simplicity. So, the final expression for DOS contains the parameter Γ which takes care of quasi particle scattering rate and can be connected with the experimental energy resolution of the ARPES data. The density of state $N_{AA}(\omega)$ as a function of various model parameters can be analyzed using numerical computation and comparison with existing results may be made.

$$N_{AA}(\omega) = \frac{1}{\pi^2}\iint_{-\pi}^{\pi}\lim_{\Gamma\to 0}\left[\frac{\Gamma}{\Gamma^2+(\omega-\mu_{1k})^2}\right]dk_x dk_y + \frac{1}{\pi^2}\iint_{-\pi}^{\pi}\lim_{\Gamma\to 0}\left[\frac{\Gamma}{\Gamma^2+(\omega-\mu_{2k})^2}\right]dk_x dk_y + \frac{1}{\pi^2}\iint_{-\pi}^{\pi}\lim_{\Gamma\to 0}\left[\frac{\Gamma}{\Gamma^2+(\omega-\mu_{3k})^2}\right]dk_x dk_y + \frac{1}{\pi^2}\iint_{-\pi}^{\pi}\lim_{\Gamma\to 0}\left[\frac{\Gamma}{\Gamma^2+(\omega-\mu_{4k})^2}\right]dk_x dk_y \quad (12)$$

3. RESULT AND DISCUSSION

3.1 IN ABSENCE OF NNN INTERLAYER COUPLING PARAMETER (t_p')

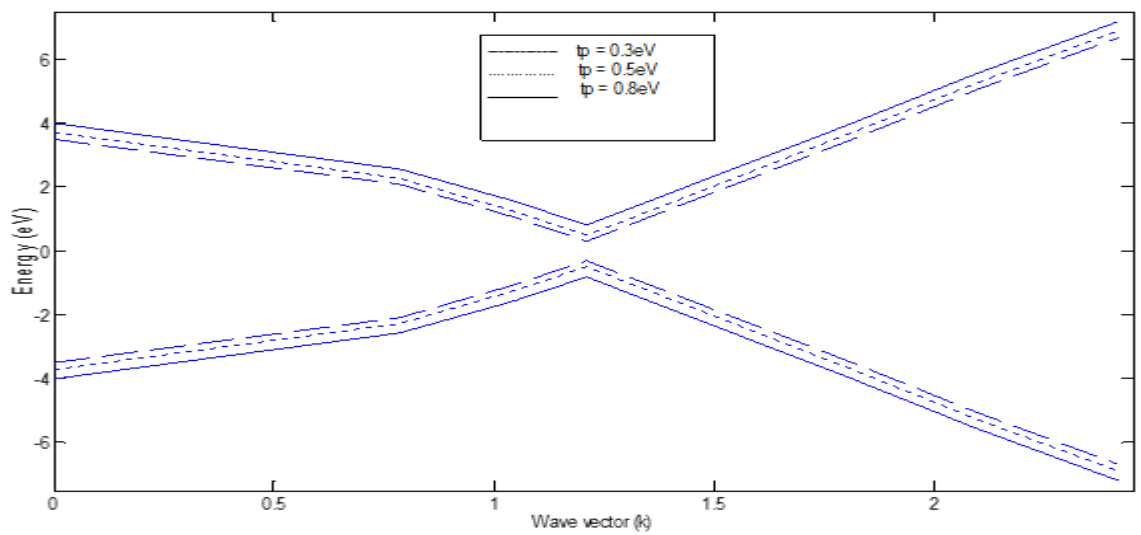

Fig. 2a Quasi particle energy vs Wave vector (t=2.6eV, t'=0.3eV, t_p = 0.3eV, 0.5eV, 0.8eV, $t_p' = 0eV$) for AA-stacked bilayer graphene.

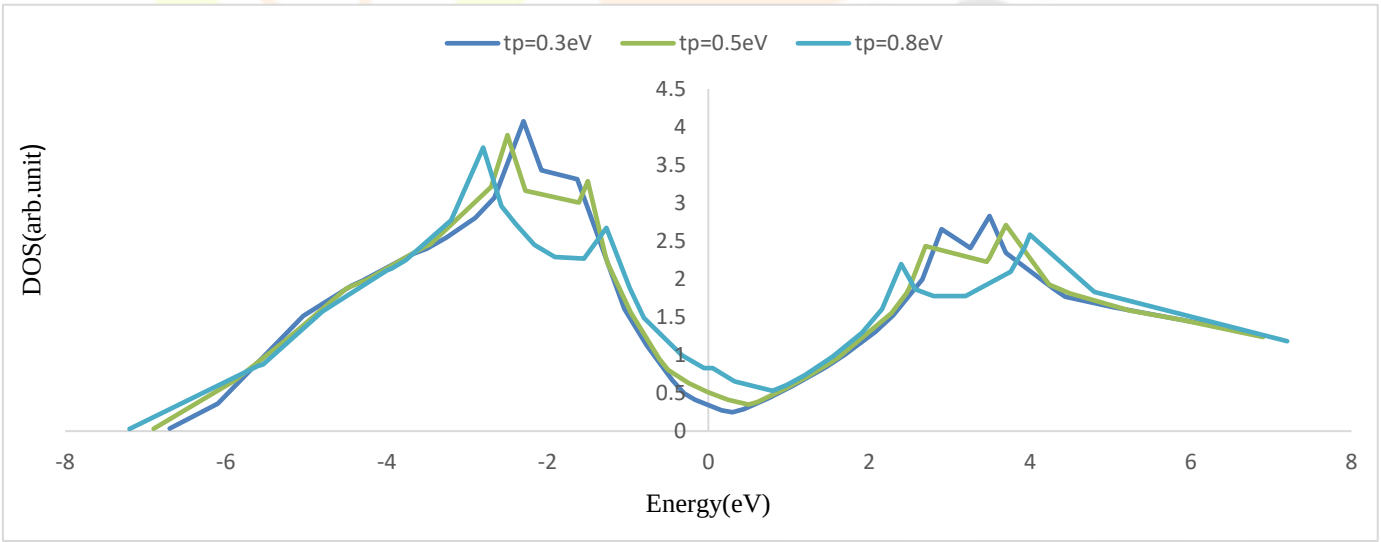

Fig. 2b Density of state vs energy (t=2.6eV, t′=0.3eV, t_p = 0.3eV, 0.5eV, 0.8eV, t_p' = 0eV) for AA-stacked bilayer graphene.

Comparision of theoretically calculated four quasi particle energies (eq. 9) and density of states (eq. 12) of AA-BLG with available experimental results is done by numerically analysing the variation of quasi particle energies with wave vector *(k)* close to Dirac point $K\left(\frac{2\pi}{3}, \frac{2\pi}{3\sqrt{3}}\right)$ within the first brillouin zone. During the numerical computation the various model parameters have been taken (t=2.6eV, t′=0.3eV, t_p = 0.3 – 0.8 eV and $t_p' = 0eV$) from recent band structure calculations and ARPES measurements relevant for AA-BLG [2,4,13]. The various model parameters have been taken (t=2.6eV, t'=0.3eV, t_p= 0.2 – 0.7 eV and $t_p' = 0eV$) from recent band structure calculations and ARPES measurements relevant for AA-BLG [2,4,7]. Figure 2(a) shows variation of quasi particle energies with wave vector k for AA-BLG. The curve shows a parabolic tunable gap between conduction and valence band at Dirac point in absence of NNN interlayer coupling parameter ($t_p' = 0$eV).Figure 2(b) shows the variation of density of states for various values of NN interlayer coupling (t_p = 0.3eV, 0.5eV, 0.8eV). It is clear from the curve that increasing values of t_p results in splitting of quasi particle peaks. Similar result has also been reported by others [4]. Graph also shows electron-hole symmetry for smaller value of t_p (0.3eV) but for higher value of t_p (0.8eV) graph breaks the particle hole symmetry as predicted by others [19] as well. Thus tunning NN interlayer coupling (t_p) via doping or gating, can result in tunable bandgap for suitable electronic properties.

3.2 IN PRESENCE OF NNN INTERLAYER COUPLING PARAMETER (*tp'*)

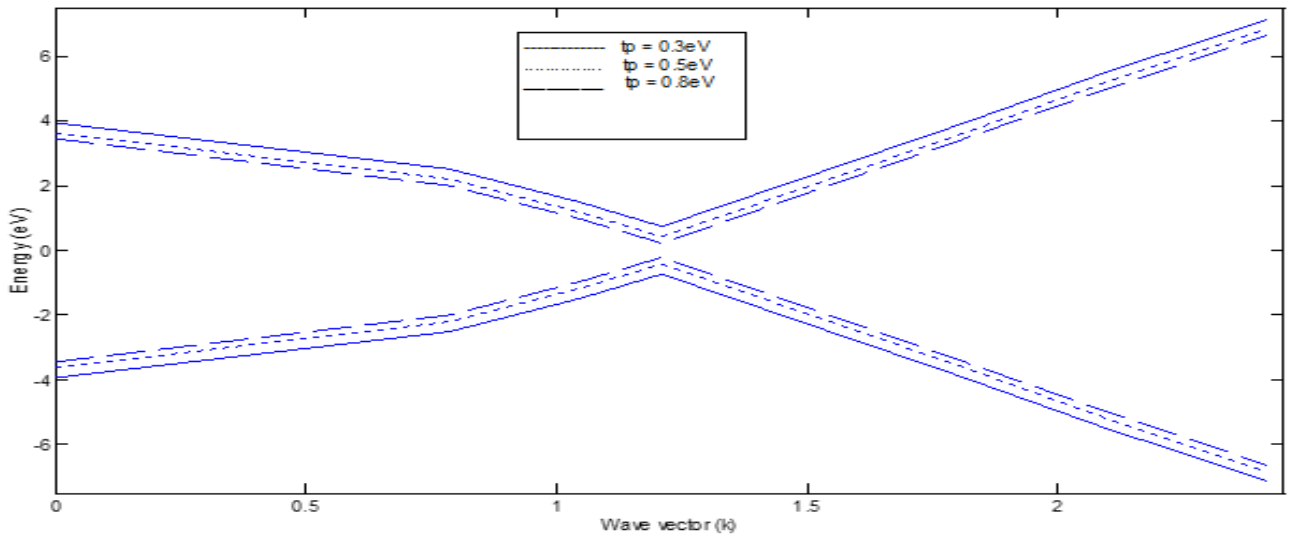

Fig. 3a Quasi particle energy vs Wave vector (t=2.6eV, t′=0.3eV, $t_p = 0.3$eV, 0.5eV, 0.8eV, $t_p' = 0.07$eV) for AA-stacked bilayer graphene.

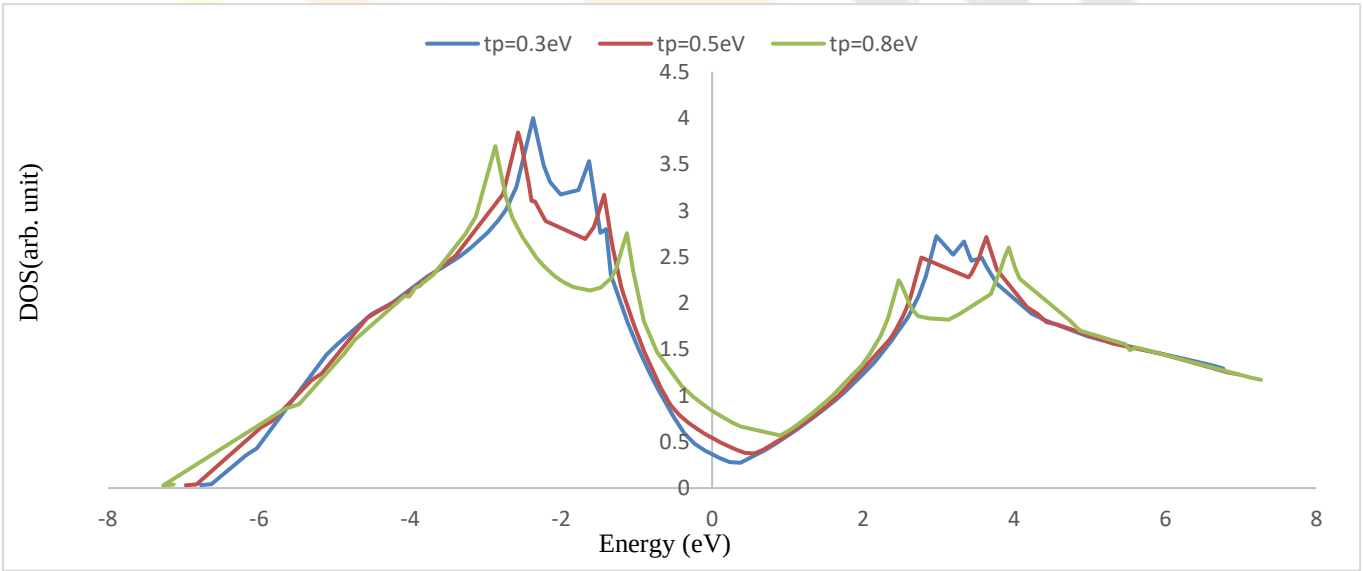

Fig. 3b Density of state vs energy (t=2.6eV, t′=0.3eV, $t_p = 0.3$eV, 0.5eV, 0.8eV, $t_p' = 0.07$eV) for AA-stacked bilayer graphene.

Our results are presented using a combination of graphs to illustrate the impact of NNN interlayer coupling on the electronic properties of AA-BLG. Figure 3(a) shows the behaviour of the quasi-particle spectrum for various values of NN interlayer coupling parameter (t_p) for fixed value of NNN interlayer coupling parameter (t_p'=0.07eV) as the values of t_p increases, energy gap between the C.B. and V.B. also increases. This opening of band gap makes AA-BLG suitable for electronic application and thus the study of interlayer interactions is crucial.

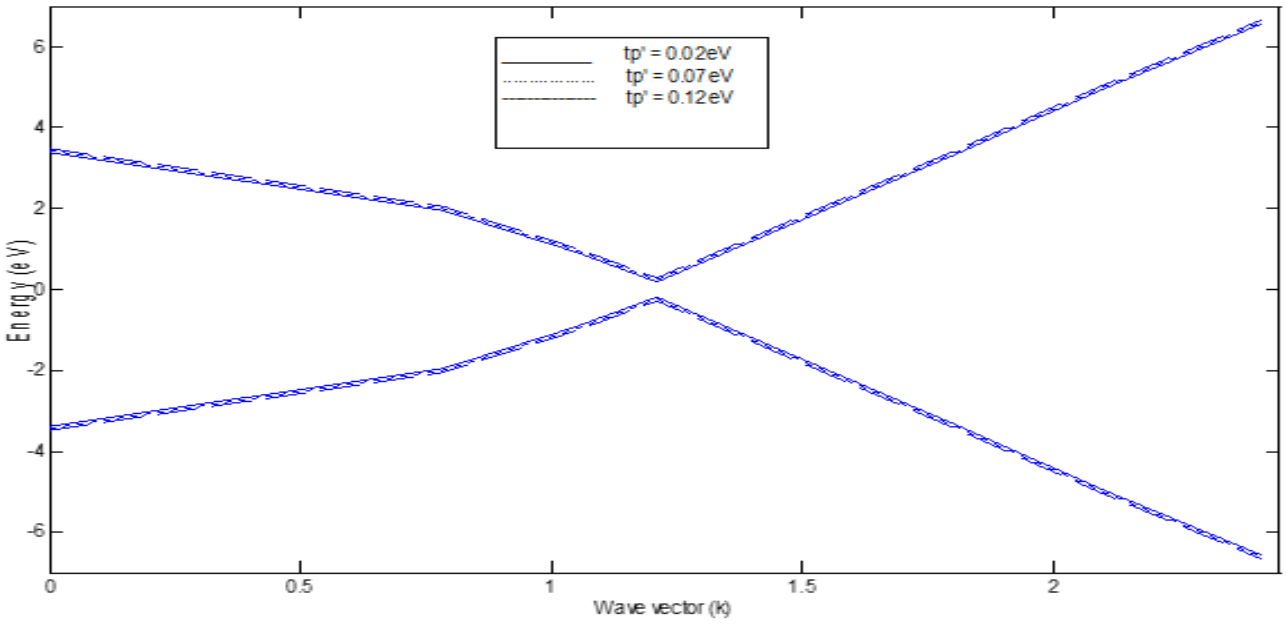

Fig. 3c Quasi particle energy vs Wave vector (t=2.6eV, t′=0.3eV, $t_p = 0.3$eV, t_p'=0.02eV, 0.07eV, 0.12eV) for AA-stacked bilayer graphene

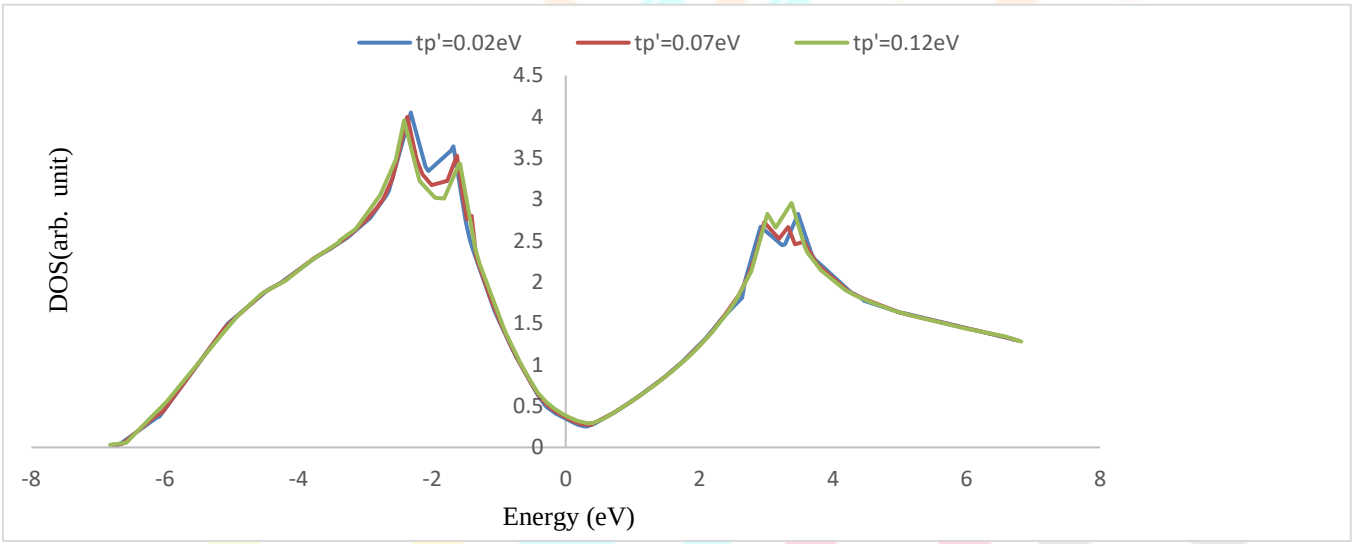

Fig. 3d Density of state vs energy (t=2.6eV, t′=0.3eV, $t_p = 0.3$eV, t_p'=0.02eV, 0.07eV, 0.12eV) for AA-stacked bilayer graphene

Figure 3(b) displays DOS versus energy plots for different nearest-neighbour coupling values (t_p = 0.3, 0.5 and 0.8 eV) while keeping the NNN coupling constant, clearly showing that an increase in t_p widens the energy gap between the conduction and valence bands. Also, the DOS peak becomes broader and show more saddle points and shorter for higher value of t_p as proposed by other researchers [2,4] as well. Figure 3(c) shows the influence of NNN interlayer coupling parameter on quasi-particle spectrum. The graph shows as t_p' increases, the energy gap reduces for fixed value of t_p(0.3eV). Figure 3(d) further demonstrates how variations in the NNN coupling parameter (t_p') affect the energy gap and shift the DOS between the valence and conduction bands. Together, these data presentations provide a detailed understanding of how both NN and NNN interlayer interactions govern the quasi-particle spectrum and overall conductivity of bilayer graphene. As t_p increases, the energy gap between the conduction and valence bands widens, indicating that stronger NN coupling enhances band splitting. This trend is especially evident in AA-BLG, where increased t_p leads to a more pronounced separation of electronic states, effectively tuning the semiconductor properties. Conversely, as the NNN coupling parameter (t_p') increases, it tends to reduce the band gap and shifts the density of states (DOS) from the valence to the conduction band, counteracting some effects of t_p. This interplay results in a more complex and tunable electronic structure where the net conductivity can be optimized by balancing both NN and NNN interlayer interactions.

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